

# Supersolid phase in spin dimer XXZ systems under magnetic field

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Using quantum Monte Carlo method, we study, under external magnetic fields, the ground state phase diagram of the two-dimensional spin  $S=1/2$  dimer model with an anisotropic intra-plane antiferromagnetic coupling. With the anisotropy  $4 \gtrsim \Delta \gtrsim 3$ , a supersolid phase characterized by a non-uniform bose condensate density that breaks translational symmetry is found. The rich phase diagram also contains a checkerboard solid and two different types of superfluid phase formed by  $S_z = +1$  and  $S_z = 0$  spin triplets, with finite staggered magnetization in z-axis and in-plane direction, respectively. As we show, the model can be realized as a consequence of including the next nearest neighbor coupling among dimers and our results suggest that spin dimer systems may be an ideal model system to study the supersolid phase.

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Five decades ago, Penrose and Onsager [1] showed that the coexistence of both off-diagonal long-range order (ODLRO) and diagonal long range order (DLRO), i.e. a supersolid (SS) state, is impossible in commensurate solids. However, it was later demonstrated [2] that vacancies in solid may still exhibit Bose-Einstein condensation. And more recently, Prokof'ev and Svistunov [3] provide a proof that the presence of vacancies and/or interstitial atoms is a necessary condition for a SS. They therefore exclude the possibility of a bulk SS in the recent experimental observations of nonclassical moment of inertia in solid  $^4\text{He}$  [4]. Instead, interpretation based on superfluid interfaces is suggested. While experimental evidence of SS is still inconclusive, numerical stimulations have already confirmed the presence of SS of bosons in various conditions, that is including next nearest neighbor interactions (n.n.n.) [5] or with softcore [6] in square lattices, and with hardcore in triangular lattices [7].

All of the models discussed above concentrate on studying bosons. Knowing the possibility of representing bosons by spins or vice versa, we wonder if SS phase also exists in spin systems. Recent experiments on spin dimer compounds, such as  $\text{TiCuCl}_3$ ,  $\text{KCuCl}_6$  and  $\text{BaSiCu}_2\text{O}_6$  [8], have confirmed the formation of super fluid (SF) of the spin triplets under an external magnetic field. These spin bosons have also previously been found to be crystallized, signaled by the magnetization plateau, in dimer compounds such as  $\text{SrCu}_2(\text{BO}_3)_2$  [9]. The competition between repulsive interaction and kinetic motion decides whether the ground state is crystalized or is a SF [10]. This also raises the possibility of a SS state. While SS is unstable against phase separation for hardcore boson in a square lattice [6, 11], the spin bosons are shown to be semi-hardcore [12]. In this work, using quantum Monte Carlo (QMC) method we demonstrate that the ground state of a spin dimer XXZ model could be SF of two kinds of triplets, a quantum solid or, most importantly, a SS. This XXZ anisotropy can be easily realized if the n.n.n. interaction among dimers is considered. There-

fore, while previous proposal of detecting SS in optical lattices [7] is technically difficult, we suggest that, due to the semi-hardcore nature of spin bosons, it may be more natural to look for a SS in spin dimer systems. Furthermore, we provide strong evidence that in the SS phase, the SF condensate amplitude has a spatial modulation and possesses a lattice symmetry on its own. This kind of non-uniform condensate has been discussed related to solid  $^4\text{He}$  [13] but has not been confirmed in numerical studies before.

In particular, we study the 2D bilayer AF spin dimer XXZ model

$$H_{XXZ} = -h \sum_{\alpha,i} S_{\alpha,i}^z + J \sum_i \mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i} + J' \sum_{\alpha,\langle i,j \rangle} (S_{\alpha,i}^x S_{\alpha,j}^x + S_{\alpha,i}^y S_{\alpha,j}^y + \Delta S_{\alpha,i}^z S_{\alpha,j}^z), \quad (1)$$

where  $\alpha = 1, 2$  denotes the layer index and the third term sums over all nearest neighbors (n.n.) in the  $xy$  plane for both layers. Since  $J \gg J'$  (we take  $J'/J = 0.29$  in this paper), the low energy physics is conveniently described by the interaction of the four spin states, singlet ( $|s\rangle$ ) and triplets  $S^z = 0, \pm 1$  ( $|t_{0,\pm}\rangle$ ) of each dimer.

When  $\Delta = 1$ , i.e., the isotropic case, the model successfully describes the SF phase, characterized by the in-plane staggered magnetization  $m_{xy}$ , of the spin  $1/2$  dimer compounds  $\text{TiCuCl}_3$ ,  $\text{KCuCl}_6$ , and  $\text{BaSiCu}_2\text{O}_6$  [12, 14]. The mean field (MF) ground state is

$$|\Psi_0\rangle = \prod_i [u|s\rangle_i + (-1)^i (v e^{i\theta} |t_+\rangle_i + w e^{-i\theta} |t_-\rangle_i)] \quad (2)$$

where  $u^2 + v^2 + w^2 = 1$  and we have neglected  $|t_0\rangle$  states here. The order parameter is given by  $m_{xy} = u(v + w)$  whose square is shown to be equal to the condensate density ( $n_0$ , the zero momentum number density) of the semi-hardcore boson  $b_i^\dagger = \frac{(-1)^i}{\sqrt{2}} (S_{1,i}^+ - S_{2,i}^+)$ , which allows up to double occupancies on each site. The global phase  $\theta$  corresponds to the orientation of the  $m_{xy}$  in the

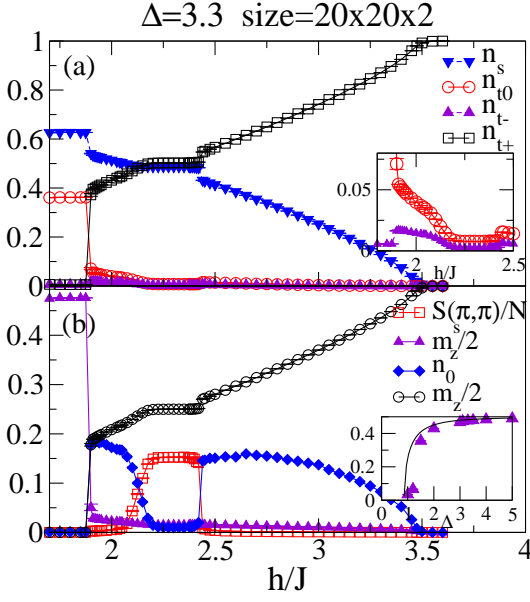


FIG. 1: (color online) Phase diagrams of (a) the number densities  $n_s$ ,  $n_{t+}$ ,  $n_{t0}$ , and  $n_{t-}$ , and (b) the order parameters  $n_0$ ,  $S(\pi, \pi)/N$ ,  $m_z/2$ , and  $m_z^s/2$ . The upper inset shows the details of  $n_{t-}$  and  $n_{t0}$  around the SS phase with enlarged scale. The lower inset compares the QMC (symbols) and the MF (solid lines) results of  $m_z^s/2$  as a function of  $\Delta$ .

$xy$  plane. Ignoring  $|t_- \rangle$ , i.e. let  $w = 0$ , it is a hardcore model with n.n. repulsive interaction  $V$  and hopping  $t$ . For the case of square lattice,  $V = t = J/2$ . This Hamiltonian preserves the particle-hole symmetry, which is broken when  $|t_- \rangle$  is taken into account and thus an asymmetric phase diagram of  $m_{xy}(h)$  is observed.

By increasing  $\Delta > 1$ , the repulsive interaction between the triplets enhances. The ratio of interaction energy and kinetic energy is determined by  $\Delta = V/t$ . Therefore, a large value of  $\Delta$  will lead to a solid phase where the structure factor has a finite peak at certain wave vector. The compound  $\text{SrCu}_2(\text{BO}_3)_2$  [9] has shown such solid phase in its magnetization plateaus. When both interaction energy and kinetic energy play crucial roles, the coexistence of both SF ordering and solid ordering is possible. Our numerical simulation indeed shows the presence of SS phase from  $\Delta \approx 3$  to 4. In our QMC simulation, we employ the stochastic series expansion approach [15] on a 2D bilayer square lattice for different linear sizes  $L = 10, 12, 16$  and 20, with temperature  $T$  inversely proportional to  $L$  such that at  $L = 20$ ,  $T/J = 0.02154$ .

In Fig.1, we show the number densities (1a) and order parameters (1b) as a function of magnetic field  $h$  for  $\Delta = 3.3$ . Let us discuss the different phases starting from low field  $h$ .

*superfluid I (SFI)* – At low field, the phase is characterized by the only non-vanishing order parameter, the staggered magnetization along  $z$ -axis  $m_z^s = \langle \sum_i (-1)^i (S_{1i}^z - S_{2i}^z) \rangle / N$  ( $N$  is the number of dimers), which is field in-

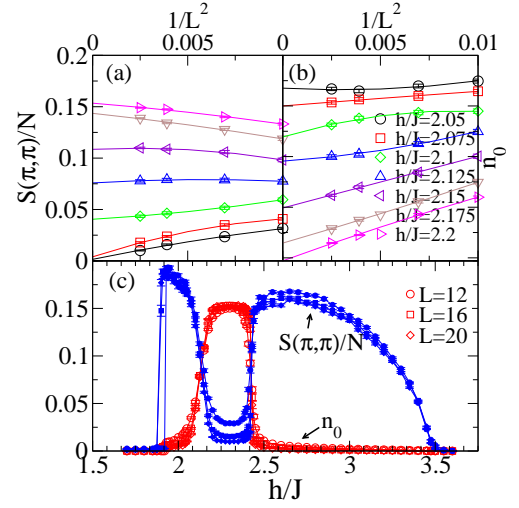


FIG. 2: (color online) Finite size scaling of (a)  $S(\pi, \pi)/N$  and (b)  $n_0$ . In (c), results of  $S(\pi, \pi)/N$  (open symbols) and  $n_0$  (filled symbols) for  $L = 12, 16$  and 20.

dependent. This is a SF phase forming by  $|t_0 \rangle$  state in the background of  $|s \rangle$  state with a MF wavefunction  $|\Psi_{t0}\rangle = \sum_i (u_0 |s\rangle_i + e^{i\phi} (-1)^i v_0 |t_0\rangle_i)$ , where  $u_0$  and  $v_0$  are given by minimizing the MF energy and have the form  $u_0^2 = \frac{1}{2}(1 + \frac{J}{4\Delta J'})$  and  $v_0^2 = \frac{1}{2}(1 - \frac{J}{4\Delta J'})$ . Then,  $m_z^s = 2u_0v_0 \cos \phi = \langle b_0^\dagger \rangle + \langle b_0 \rangle$ , with  $b_0^\dagger$  is the hardcore boson operator that creates a  $|t_0 \rangle$  state. At finite field, the global phase  $\phi$  is fixed to zero so that the largest  $m_z^s$  aligned to the field direction. The inset of Fig.1b shows an excellent agreement of MF  $m_z^s/2$  to the values obtained from QMC for different  $\Delta$ 's. Even for  $\Delta = 1$ , that is relevant to many spin dimer materials, as long as  $1 > J'/J > 1/4$ , so  $v_0 > 0$ , there is still a small but finite  $m_z^s$  in this low field phase.

*Superfluid II (SFII)* – When  $h/J \gtrsim 1.87$ , the strong Zeeman energy favors  $|t_+ \rangle$  instead of  $|t_0 \rangle$  and leads to the condensation of  $|t_+ \rangle$ . In Fig.1(b), the condensate density  $n_0 = m_{xy}^2$  of boson  $b^+$ , jumps abruptly to about 0.2 as  $m_z^s$  drops to a small value. Note that  $n_{t0}$  is small but still finite so that  $|t_0 \rangle$  behaves as impurities. This phase reappears at even higher fields.

*Supersolid (SS)* – At  $h/J \sim 2.10$ ,  $n_0$  begins to reduce rapidly while the structure factor  $S(\mathbf{Q})/N = \sum_{ij} \langle n_{i,t+} n_{j,t+} e^{i\mathbf{Q} \cdot \mathbf{r}_{ij}} \rangle / N^2$  at the wave vector  $(\pi, \pi)$  of  $|t_+ \rangle$  increases. The single peak of  $S(\pi, \pi)/N$  implies a checkerboard solid ordering of  $|t_+ \rangle$  in the system. The transition from SFII to SS is also signaled by the kinks observed in the  $m_z$ ,  $n_0$ ,  $n_{t-}$ , and  $n_{t0}$  (Fig.1). Within  $2.2 \gtrsim h/J \gtrsim 2.1$ , both SF and solid order parameters remain finite and constitute the SS phase. Finite size scaling shown in Fig.2 indicates that the SS phases is stable in the thermodynamic limit. To address the concerns of phase separation, i.e. the possibility of having separate domains of checkerboard solid and SF, we measure the

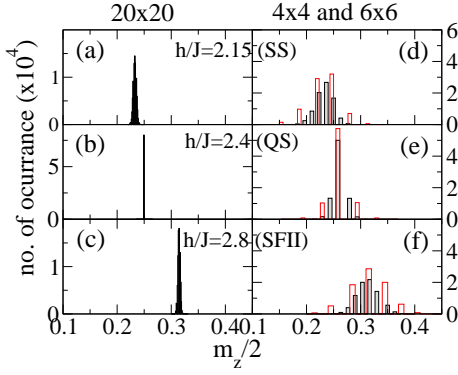


FIG. 3: (color online) Histograms of the averaged  $m_z/2$  of the whole lattice ((a) to (c)) and of a small area ((d) to (f)) of size 4x4 (unshaded) and 6x6 (shaded).  $\Delta = 3.5$ . For comparison, histograms of SFII and QS are also presented.

average  $m_z$  within a small area of size 6x6 and 4x4 of a 20x20 lattice. Since  $m_z$  differs for different domains, the small area measurement should give two distinct peaks in the histogram in the case of phase separation. Shown in Fig.3 are the histograms of  $m_z/2$  for selected fields. The distribution is wider in 6x6 case when compared to that of whole 20x20 lattice where fluctuation is averaged out. The single Gaussian peak at  $h/J = 2.15$  indicates that there is no domains of different magnetization and rules out the possibility of phase separation.

**Quantum solid (QS)** – Increasing the field further ( $h/J > 2.20$ ) will half-fill the system with  $b^+$  so that  $m_z = 0.5$ . Because of strong repulsive interaction between nearest neighbor  $b^+$ , adding or removing extra boson to this checkerboard solid state requires a finite energy. This causes the stability of the state and the plateau in the  $m_z$  curve. Differing from the classical value  $S(\pi, \pi)/N = 1/4$ , the calculated structure factor in the solid phase is about 0.13 for  $\Delta = 3.3$ . The  $m_z$  of each sublattice are found to be 0.890 and 0.110 respectively (Table I). As  $\Delta$  increases,  $S(\pi, \pi)/N$  approaches the classical value of 1/4. At higher fields, the melting of the solid is of first order and the SFII phase reappears.  $n_{t+}$  keeps increasing while all other states reduces to zero at  $h = 3.50$ , where all the spins are fully polarized.

Here we address the question of non-uniform condensate in the SS phase. Table I shows that the  $m_z$  differs in different sublattices A and B, and breaks the translation symmetry. To examine the condensate more closely, we define the sublattice condensates  $n_A$  and  $n_B$  as  $n_\alpha = \sum_{i \in \alpha} \langle b_i^\dagger \rangle / N$ , where  $\alpha = A, B$ . In our simulation, we compute the products of sublattice condensate  $n_{\alpha\beta} = \sum_{i \in \alpha, j \in \beta} \langle b_i^\dagger b_j \rangle / N^2$ . As shown in Table I,  $n_{AA} \neq n_{BB}$  and  $n_{AB} = n_{BA} = \sqrt{n_{AA}n_{BB}}$  in the SS phase. Thus  $n_{\alpha\beta} = n_\alpha n_\beta$ , and  $n_A = 0.130$ ,  $n_B = 0.148$ . It indicates that the condensate has a checkerboard ordering on its own, which is the intrinsic nature of a SS state [13]. This is different from the idea of having a SF

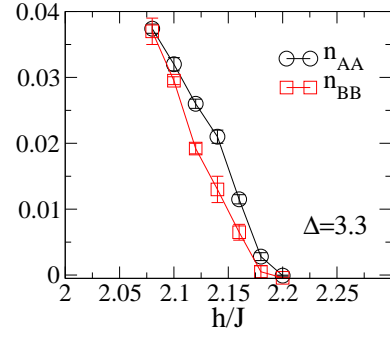


FIG. 4: (color online) Sublattice condensates  $n_{AA}$  and  $n_{BB}$  in the SS phase extrapolated to  $T \rightarrow 0$  and  $L \rightarrow \infty$ .

component "on top" of a bose solid. The checkerboard order of the condensate disappears in the usual superfluid phase SFII, while the condensate amplitude on both sublattices vanishes in the QS phase as expected. The result for different  $h$  is plotted in Fig. 4. The SS phase, instead of being a crossover between SFII and QS, is clearly a new phase with second order phase transitions from SFII and QS respectively. Nevertheless, a more detail study of scaling behavior is needed to determine the scaling exponents. This result also rules out phase separation.

Fig.5(a) and (b) show the plots of order parameters as a function of  $h$  for different  $\Delta$  and the phase diagram of  $\Delta$  vs.  $h$ , respectively. The isotropic case,  $\Delta = 1$ , is similar to those observed experimentally on other spin dimer compounds. Enhancing the anisotropy to  $\Delta = 3$ , there is a small drop in  $n_0$  around  $m_z = 0.5$  while  $S(\pi, \pi)/N$  remains zero. For even larger repulsive interaction at  $\Delta = 3.1$ , the drop develops further with a finite peak of  $S(\pi, \pi)/N$  at  $m_z = 0.5$ . Note that it is a commensurate SS that is made possible by the semi-hard core nature of the spin bosons. The finite peak of  $S(\pi, \pi)/N$  finally develops to a plateau together with the  $m_z$  in the QS phase. As a consequence of lowering energy of  $|t_0\rangle$  state with increasing  $\Delta$ , the phase boundary of SFI to SFII moves to the higher fields and shrinks the region of SFII and SS which eventually vanishes at  $\Delta \sim 4$  and leaves a first order transition from SFI to QS phase. As shown in Fig.5(b), the SS phase is stable in a parameter region with  $4 \gtrsim \Delta \gtrsim 3$ .

The next question is whether these phases can be re-

	$m_z^A/2$	$m_z^B/2$	$n_{AA}$	$n_{BB}$	$n_{AB}$	$n_{BA}$
SFII	0.224(5)	0.225(5)	0.0384(7)	0.0384(6)	0.0384(7)	0.0387(6)
SS	0.397(1)	0.0831(9)	0.0169(5)	0.0219(5)	0.0195(5)	0.0194(4)
QS	0.4449(2)	0.0549(2)	0.0017(1)	0.0036(1)	0.0026(1)	0.0027(1)

TABLE I: Sublattice magnetizations and condensates for SFII ( $h/J=2.08$ ), SS ( $h/J=2.14$ ) and QS ( $h/J=2.24$ ) phases.  $\Delta = 3.3$  and  $L=20$ . The small values of  $n_{\alpha\beta}$  in QS phase is due to the finite size effect.

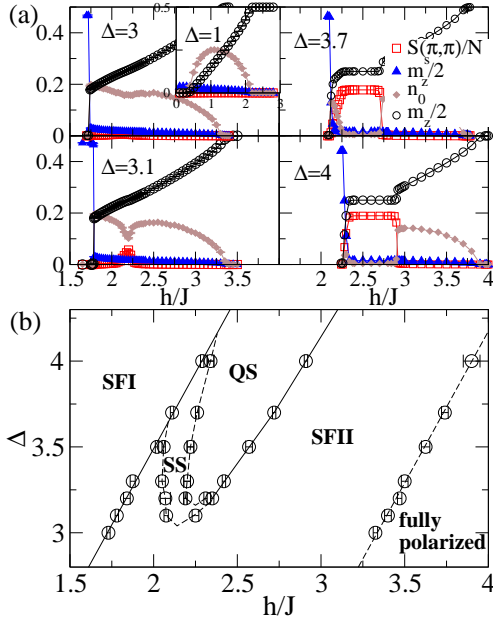


FIG. 5: (color online) (a) Order parameters as a function of  $h/J$  for different  $\Delta$  with lattice size  $20 \times 20 \times 2$ . The inset shows the isotropic case ( $\Delta = 1$ ). (b) Ground state phase diagram (extrapolated to  $L \rightarrow \infty$ ) of  $\Delta$  vs.  $h/J$ . Solid (dotted) lines denote the first (second) order phase boundaries.

alized in real material. There are two issues, however, that have to be considered. The first is the anisotropy that introduced in  $H_{XXZ}$  only applies to the interdimer coupling but not the intradimer coupling. This kind of selective anisotropy is hard to realize. The second issue concerns the large value of  $\Delta \sim 3$  for the SS to be stable. In usual compounds the XXZ anisotropy is caused by the spin-orbit coupling, which couples the spin to the crystal structure, and the effective  $\Delta$  is much smaller than 3. To the first issue, one may instead consider a  $S = 1$  compound with a single ion anisotropy  $D \sum (S^z)^2$ , where  $D$  plays the role of intra-dimer coupling  $J$  and the states  $S^z = \pm 1$  and  $S^z = 0$  plays the role of  $|t_{\pm}\rangle$ , and  $|s\rangle$ , respectively. There is no corresponding states of  $|t_0\rangle$ . Therefore, SFI is replaced by  $S^z = 0$  state on each site. Together with spin-orbit coupling that realizes the XXZ anisotropy, one has essentially the same phase diagram as in the case of  $H_{XXZ}$ , although a large enough anisotropy is still needed. On the other hand, there is another route to look for the realization of the SS phase. Consider two neighboring  $t_+$  states, the inclusion of n.n.n. coupling  $J'' \mathbf{S}_{1,i} \mathbf{S}_{2,j}$  ( $i, j$  are n.n.) will simply enhance the repulsion between  $|t_+\rangle$  states. Contrastingly, if  $|t_0\rangle$  is ignored,  $J''$  reduces the hopping between the neighboring  $|t_+\rangle$  and  $|s\rangle$  states due to the antisymmetry of  $|s\rangle$ . In bond operator representation, when the  $|t_0\rangle$  states is ignored, spin operators  $S_{1,2}^z \approx \frac{1}{2}(t_+^\dagger t_+ - t_-^\dagger t_-)$  and  $S_{1,2}^\pm \approx \pm \frac{1}{\sqrt{2}}(s^\dagger t_- - t_+^\dagger s)$ . Then the n.n.n. coupling

is approximately given by

$$\mathbf{S}_{1,i} \mathbf{S}_{2,j} \approx S_{1,i}^z S_{1,j}^z - \frac{1}{2}(S_{1,i}^+ S_{1,j}^- + S_{1,i}^- S_{1,j}^+). \quad (3)$$

The inclusion of  $J''$  now breaks the spin rotational symmetry and leads to an XXZ model with effective n.n. coupling of  $J^* = J' - J''$  and anisotropy  $\Delta$  given by  $J^* \Delta = J' + J''$ . Therefore, if  $J'' = J'/2$  one has a spin dimer XXZ model with  $\Delta = 3$ , close to the SS phase.

In summary, we demonstrate that the spin dimer XXZ model, a natural semi-hardcore boson system with defects, contains a SS phase that characterized by the bipartite condensate density. The anisotropy can be a consequence of including the n.n.n. coupling among dimers. While it is suggested [7] that SS state of hardcore bosons can be tested in triangular optical lattices, we propose that spin dimer compounds may be a natural place to realize the SS state of semi-hardcore bosons.

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